Mostovoy Reply: In their Comment [1] Kenzelmann and Harris argue against the conclusion made in [2] that spiral magnets are in general ferroelectric. First of all, I believe, this conclusion was proved experimentally. The systematic search for ferroelectricity in magnets with spiral ordering recently led to a discovery of new multiferroic materials, such as $CoCr_2O_4$ [3], $MnWO_4$ [4, 5] and $LiCu_2O_2$ [6].

Furthermore, Kenzelmann and Harris argue that the continuum theory outlined in [2] leads to *misleading predictions* about the magnetically-induced electric polarization. To prove their point, they consider two hypothetical spin configurations shown in Fig. 1 (c) and (d) of their Comment, and argue that the results of the continuum theory are incompatible with crystal symmetries. While one cannot deny the importance of symmetry considerations, the arguments Kenzelmann and Harris are themselves very misleading. They incorrectly assert that for the spin configurations shown in Fig. 1 (c) and (d) 'the spiral theory' would predict electric polarization along, respectively, the c and a axes.

The continuum model of multiferroics [2] is based on assumption that the spin state can be described by a single magnetization vector. For $TbMnO_3$ (see Fig. 1b), where the wave vector of the magnetic spiral is along the b axis and spins are rotating in the bc plane, it predicts electric polarization $\mathbf{P}$ along the c axis, in agreement with experiment. The magnetic structures (c) and (d) are of a different kind, as they are made of spirals rotating in opposite directions. Thus in the configuration (c) there are two counter-rotating bc spirals in each ab plane, which is why the net polarization along the c axis is zero. Similarly, in the configuration (d) the ab spirals in neighboring bc planes rotate in opposite directions, resulting in zero net P_a.

It is not difficult to modify the continuum model considered in [2] to describe these more general magnetic orders. For more than one magnetic ion per unit cell one can introduce several independent magnetic order parameters, which increases the number of possible magnetoelectric coupling terms. For instance, all three spin configurations shown in Fig. 1 of the Comment can be described by three antiferromagnetic order parameters

$$\begin{aligned} \mathbf{L}_1 &= \mathbf{S}_1 + \mathbf{S}_2 - \mathbf{S}_3 - \mathbf{S}_4, \\ \mathbf{L}_2 &= \mathbf{S}_1 - \mathbf{S}_2 + \mathbf{S}_3 - \mathbf{S}_4, \\ \mathbf{L}_3 &= \mathbf{S}_1 - \mathbf{S}_2 - \mathbf{S}_3 + \mathbf{S}_4 \end{aligned} \tag{1}$$

(the labels of the 4 Mn ions in the unit cell of $TbMnO_3$ are the same as in [7]). The spiral configuration (b) can be described by a single order parameter $\mathbf{L}_1$ with nonzero L_1^b and L_1^c. As discussed in [2], the magnetoelectric coupling linear in the gradient of the magnetic order parameter (Lifshitz invariant) allowed by symmetries has the form $P^c\left(L_1^c\partial_y L_1^b - L_1^b\partial_y L_1^c\right)$, which gives rise to magnetically-induced P^c. The configuration (c) is described by two different order parameters, L_1^b and L_3^c. The term $L_3^c\partial_y L_1^b - L_1^b\partial_y L_3^c$ does not transform like any of the components of $\mathbf{P}$, so that the induced polarization is zero. Finally, for the configuration (d) with nonzero L_1^b and L_2^a, the only possible coupling term is $P^c\left(L_1^b\partial_y L_2^a - L_2^a\partial_y L_1^b\right)$, allowing for nonzero P^c.

The point is, however, that the spin configurations (c) and (d) considered by Kenzelmann and Harris, are very artificial, as it is difficult to find a system where interactions between spins would favor the simultaneous presence of counter-rotating spirals. The average interaction between counter-rotating spirals is zero, while for spirals with spins rotating in the same direction some interaction energy can always be gained by properly adjusting their relative phases. This is the reason why the simple model of Ref. [2] with a single vector order parameter successfully describes thermodynamics and magnetoelectric properties of many spiral multiferroics.

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[1] M. Kenzelmann and A. B. Harris, Comment arXiv/cond-mat0610471.
[2] M. Mostovoy, Phys. Rev. Lett. **96**, 067601 (2006).
[3] Y. Yamasaki, S. Miyasaka, Y. Kaneko, J.-P. He, T. Arima, and Y. Tokura, Phys. Rev. Lett. **96**, 207204 (2006).
[4] K. Taniguchi, N. Abe, T. Takenobu, Y. Iwasa, and T. Arima, Phys. Rev. Lett. **97**, 097203 (2006).
[5] O. Heyer *et al.*, J. Phys. Condens. Matter **18**, L471 (2006).
[6] S. Park, Y. J. Choi, C. L. Zhang and S.-W. Cheong, to be published.
[7] A. B. Harris and G. Lawes, arXiv/cond-mat0508617.